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NEW OPTIONS FOR MANAGING DEPLETED URANIUM MATERIALS

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I. ABSTRACT

Uranium hexafluoride management options have been increased as a result of new technologies sponsored by the DOE Environmental Management Office of Science and Technology (OST). These technologies include an alternative uranium metal production technique, a high density depleted uranium aggregate used to make a high density concrete (DUCRETE) for radiation shielding applications, and a method for encapsulating depleted uranium oxide into polyethylene for shielding applications. Both the DUCRETE aggregate and the polyethylene can also serve as advanced waste forms for stabilizing the depleted uranium. These technologies offer additional management options for converting, using, or disposing of the large inventory of depleted uranium stored at the DOE enrichment plants.

II. INTRODUCTION

In 1993, OST began sponsorship of projects to evaluate management options for depleted uranium inventories throughout the complex. Most of the work was focused on DOE's 550,000 MT inventory of UF_6 stored at the DOE Enrichment Plants (two of which are now owned and operated by the USEC). Although the UF_6 inventory represents over 95% of the total inventory, additional large quantities of depleted uranium in various chemical forms exist at Fernald and at the Savannah River Sites. Minor quantities are located at other sites including Rocky Flats, INEEL, and LANL. OST began investigations to determine if advanced technology might reduce the future cost of depleted uranium management. A summary of the OST findings from its projects on depleted uranium is contained in Reference 1. This paper presents the most significant results from this sponsored research for management and use of the surplus depleted uranium.

While the UF_6 inventory has not been declared a waste, there was a desire to estimate the cost impact if such action were to occur. Thus, the program first evaluated disposal options and determined that there was a $3 billion to $11 billion liability for converting the UF_6 inventory into a form suitable for disposal and disposing of it at either the Nevada Test Site or at Hanford as a low-level or mixed waste (Reference 2). The mixed waste option was considered since if the depleted uranium were declared a hazardous waste in the future, it would have to be managed as a mixed waste under RCRA. From this cost baseline, technology development concepts were evaluated which would reduce the costs of conversion, which would support depleted uranium recycling, or which would reduce the cost of disposal. Since depleted uranium is a regulated radioactive material, the most feasible products were those where institutional control was easily facilitated such as nuclear shielding.

III. RESULTS

The major technologies identified that could contribute to large cost savings in the future depleted uranium recycle or disposal efforts included:

- Direct Plasma Conversion of UF_6 to U metal
- DUCRETE and Polyethylene Radiation Shielding Technology
- DUAGG Aggregate and Polyethylene Waste Form Technology

Each of these technologies is described below with the estimated cost savings. Inherently, since these studies were only carried to the bench scale stage at DOE research laboratories, there exists considerable uncertainty in the estimated baseline costs and cost savings.

A. Plasma Conversion of UF_6 to U metal

1. Technology Description

This technology was conceived at the INEEL as a possible method for a lower cost production of uranium metal from UF_6. Conceptually, the process operates as shown in Figure 1. The UF_6 is injected as a gas into the center of a hydrogen-rich plasma operating at about 5000°C. At this temperature, the UF_6 dissociates to uranium metal and elemental fluorine. The fluorine combines with the hydrogen to form HF. As the gas exits the plasma reactor, it must be cooled rapidly to avoid the back reaction of HF with uranium. The uranium metal condenses from the vapor state to a nanometer sized particle.

At the exit of the reactor, the gas (HF and unreacted H_2) with the entrained uranium metal particles enters a cyclone where the metal is separated from the gas. The system operates with excess hydrogen that must be separated from the HF gas and recycled. Consequently, with recovery of the HF from the gas stream, the process produces no waste products.

Advantages of this direct conversion approach must be contrasted with the traditional method for uranium metal production – the Ames Process. In the Ames Process, UF_6 is reacted with hydrogen to produce UF_4 or "greensalt" and HF. In a subsequent step, the greensalt is mixed with magnesium metal chips, heated in a graphite-lined retort (high temperature crucible) to about 800°C where an exothermic reaction begins producing uranium metal and MgF_2. This reaction liberates tremendous heat and the entire mass of uranium and MgF_2 become molten. Due to density differences, the U metal liquid phase separates by gravity from the MgF_2 salt to form a "derby" which takes the shape of the graphite-lined crucible. The MgF_2 floats above the uranium metal. After this material is cooled, the uranium metal derby is manually separated from the MgF_2 salt. The MgF_2 salt is disposed as a radioactive wastes as it typically contains 1% to 2% uranium by weight. The metal derby is then cleaned and alloyed as required in a vacuum casting furnace. The metal is then finished into products via extrusion, rolling or casting.

The major advantages of the INEEL process are expected to be continuous operation and reduced waste streams. The Ames process is performed as a batch process in retorts holding a 1000 to 4000 lb of UF_4. Most of the operations are labor intensive. As seen from above, the INEEL plasma conversion process consumes only hydrogen and liberates only anhydrous HF as a byproduct. The HF gas is comparably easy to purify and sell. In contrast, the greensalt reduction reaction consumes magnesium as a reactant and produces MgF_2 contaminated with uranium.

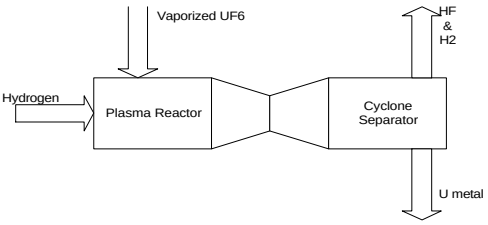

Figure 1. Plasma Reactor Functional Features

The major challenge with the plasma reduction process is associated with the design and maintenance of a plasma reactor operating at such high temperature for the conversion of UF_6 to uranium metal. INEEL researchers identified methods for designing such reactors but demonstration beyond bench scale was beyond the scope of the funded program.

2. Estimated Cost Savings

After the proof of concept bench scale test was successful, an engineering conceptual design was performed for INEEL by Morrison Knudsen to estimate the total lifecycle cost of this approach (Reference 3). In summary, this report identified that a reasonably sized facility could produce uranium metal at about $3.00 /kg. This compares to estimated commercial costs from the Ames process of about $8.80 /kg (Reference 4).

If this process were implemented and applied to the full inventory of depleted uranium, the total cost savings would be of the order of $3.19 billion (550,000 MT*1000kg/MT*[$8.8-$3.00]). This potential cost saving is of course premised upon a need for converting all of the material into uranium metal. Nevertheless, this study demonstrated a large potential cost savings associated with this advanced technology. The total cost of the research to date supporting this effort at the INEEL was approximately $250K.

B. DUCRETE Shielding Technology

1. Technology Description

Depleted uranium metal has long been used in special nuclear shielding applications where space and weight were key system performance parameters. Broader use, while technically feasible, is deterred by cost considerations. Generally, materials such as steel, lead, and concrete can be deployed as nuclear shielding at lower cost than depleted uranium metal. DUCRETE Concrete technology was developed at INEEL as an

alternate way to use uranium in a shielding application but at much lower cost than for uranium metal.

The concept converts uranium in any form (UF_6, Umetal, UO_3, and U_3O_8) into a ceramic aggregate of mostly UO_2. Conversion of UF_6 to an oxide is a well known industrial process. INEEL staff focused on using either U_3O_8 or UO_3 as the feed material for production of UO_2 aggregate. The UO_2 aggregate (DUAGG) was then be used as the large aggregate in a conventional concrete mixture to produce DUCRETE Concrete (Reference 5). Because of the exceptionally high density of the UO_2 aggregate (about 8 gm/cm^3), DUCRETE Concrete has density (5.7 to 6.0 g/cm^3) of almost 3 times that of conventional concrete (2.1 g/cm^3). Such high density will produce superior radiation shielding compared to traditional concrete (Reference 6). A US Patent was allowed on the DUCRETE concept and the associated material composition. Foreign patents have been applied for.

In general, the largest cost element of this DUCRETE Concrete concept was the conversion of UF_6 to U_3O_8 . Good cost data for conversion of UF_6 to U_3O_8 were difficult to come by as commercial companies consider this data confidential. DOE reports from Oak Ridge contained conversion cost data that could be used to produce an estimate of $8.40 /kg-U (Reference 7). Other cost data available indicated that a commercial operation could offer conversion at about $4.20 /kg-U (Reference 8).

Given that DOE needs to eventually convert the UF_6 into a more stable compound (Reference 9) regardless of costs, this conversion cost must be borne by DOE regardless of the final use of the material. New technologies being developed by Starmet and others promises significant reductions in UF_6 conversion costs. Aggregate production cost estimates that ranged between $0.25 to $0.65 cents per pound, where the plant capacity ranged from 80 to 16 tons per day, respectively.

Consequently, the deployment of DUCRETE Concrete technology clearly appeared to be a much lower cost option for using depleted uranium in shielding applications for two reasons: 1.) lower cost to produce the uranium product (DUAGG) and, 2.) lower cost to fabricate components from DUCRETE concrete compared to depleted uranium metal.

The latter point requires some additional explanation. Depleted uranium metal fabrication is done by combinations of vacuum casting, extrusion, rolling and machining to final shapes. Surveys made for DOE established a range for manufacturing costs for depleted uranium metal fabrication from $4.40 to $22 /kg-U (Reference 4). Conventional concrete, by comparison, is usually priced at something like $200 per cubic yard installed (~4000 lbs). This works out to about $0.05 per pound. Excluding the cost of the aggregate, DUCRETE concrete will have fabrication costs similar to traditional concrete. At this low price, it is clear that the fabrication cost for DUCRETE material could be many times that of conventional concrete and still be nowhere as expensive as depleted uranium metal.

2. Estimated Cost Savings

Again, the cost savings with this technology is contingent upon the deployment option selected by the DOE. However, two points are worth emphasizing:

- There are over 550,000 metric tons of UF_6 that must be converted into a more stable final form and this means conversion to U_3O_8 as a minimum. Thus, the most costly element of DUCRETE Concrete is a committed cost under all circumstances. Manufacturing and use of the aggregate into DUCRETE shielded containers is a relatively small marginal cost compared to containers from traditional concrete.
- The depleted uranium is a radioactive toxic metal. Consequently, any future use or disposal option will ultimately require a disposal facility. Thus, the lowest cost final solution should be as part of a waste package that requires disposal anyway – i.e., spent fuel and high level waste. By integrating depleted uranium management with the disposal of high level waste and spent fuel, various synergistic options become available. The most significant option is to use DUCRETE Concrete as shielded packages in the geologic repository and avoid the need for expensive remote operations for waste emplacement and monitoring for over 100 years.
- The use of depleted uranium as DUAGG in DUCRETE Concrete produces an outstanding final form that will offer maximum leach resistance where ever the material is finally disposed.

Thus, if the depleted uranium were to be converted into spent fuel storage casks and into fuel overpacks for use in Yucca Mountain, all of the depleted uranium could be used, the disposal issue would be solved, and operation of Yucca Mountain in the 100 years after fuel loading would be simpler. In addition to any projected cost savings, the repository operational safety and ease of maintenance should also be improved. A detailed impact on total capital cost has not been performed. This concept also responds to the request of the Nuclear Waste Technology Review Board for a simpler design concept for the repository.

The minimum cost saving associated with the above concept would be about $1 billion by avoiding the separate disposal of the uranium oxide as low-level waste and the associated post-burial surveillance.

C. DUCRETE HLW Casks at the SRS

1. Technology Description

A specific opportunity to deploy DUCRETE Shielding technology has been identified at the Savannah River Site where 55 million lb. of uranium oxide (UO_3) is stored. This uranium has been recovered from fuel reprocessing activities over the last 30 to 40 years. At the present time, and absent a new nuclear arms buildup program, there is no known use for this material. Consequently, only time is preventing it from officially being declared a waste. Using the DUCRETE Shielding option, a proposal and Deployment Plan (Reference 10) for conversion of this material into HLW storage casks have been prepared and submitted to DOE as Advanced Technology Deployment Program Project. This project would use both the contaminated metal from the SRS reactor heat exchangers and the depleted uranium oxide now stored at SRS for building the casks.

2. Estimated Cost Savings

Compared to the alternative baseline plan of building a Second Glass Waste Storage Building, this option saves DOE over $100 million using conservative avoided cost values for the UO_3 and heat exchanger metal disposal. The facility cost estimate for using casks versus a additional storage building also produced a savings of over $34 million. Total DOE investment in the DUCRETE technology is less than $1 million.

D. DUAGG and Polyethylene Waste Forms

1. Technology Description

The DUCRETE Technology Program developed at the INEEL under EM50 funding has produced a high-density waste form from depleted uranium oxide -- DUAGG. As discussed above, this aggregate was made for use in DUCRETE Concrete. Leaching tests at the INEEL have established the DUAGG material to have very low leach rate characteristics in the ANS 16.1 test procedure. Consequently, even if DUCRETE Concrete is never deployed, the DUAGG aggregate still represents a high quality, leach resistant waste form.

EM50 has also supported another technology developed at Brookhaven National laboratory for uranium oxide waste stabilization. This process uses polyethylene to micro encapsulate uranium oxide. Uranium oxide (UO_3) loading from 50 to 90 wt% of UO_3 were reported by P. Kalb from BNL (Reference 11). Since U_3O_8 is the normal product from UF_6 reduction, the data from Kalb had to be adjusted for use in this paper. Using his data for 90% waste loading and adjusting the measured polyethylene-UO_3 density to a predicted density for polyethylene-U_3O_8, a density of 4.2 g/cm3 was developed and used for the analysis.

2. Estimated Cost Savings

DUAGG has a density of at least 8 g/cm^3 and a packaging or bulk density of 4.8 g/cm^3 (at 60% packing density[1]). The uranium oxide loading as UO_2 in DUAGG is over 90 weight percent in the aggregate. Thus, the effective uranium oxide density in the packaged DUAGG waste form is 4.46 g/cm^3 (0.9 X 4.8). This is compared to cement grout (with a mass loading of 25%) where the effective uranium oxide (U_3O_8) density in the waste form is 0.43 g/cm^3 and the drum loading efficiency is 95%. For polyethylene, the effective loaded density was 2.23 g/cm^3 at a loading efficiency of 95%. From this comparison, it is clear that both EM50 supported technologies will provide a substantially improved waste loading compared to traditional grout technology. The consequence of this higher uranium oxide loading in polyethylene and DUAGG aggregate is lower total disposal quantity as shown in Table 1 for a stabilized uranium oxide waste form.

A model was developed to estimate the disposal cost savings of grout, polyethylene, and DUAGG aggregate. The model inherently contains many assumptions regarding the cost of transportation, disposal fee, container cost, capital and operating cost. Most of these assumptions are listed in Table 2. For equal processing cost and the assumptions chosen, the unit cost of grout, polyethylene and DUAGG are $4.28, $2.02, and $1.31 per kg-U stabilized.

Thus, the cost savings of the DUAGG waste form if applied to the disposal of the entire 550,000 MT inventory of UF_6, is $1,106 million compared to grout. The savings from the use of polyethylene micro-encapsulation, while not as large as for DUAGG aggregate, is $841 million. DUAGG Aggregate stabilization is calculated to be $265 million less than for polyethylene.

The total DOE investment at INEEL for the DUAGG and DUCRETE technology has been about $1.5 million. Investment costs at Brookhaven for the

[1] Bulk density of 58% has been measured with whole briquettes. A packing density of 70% is expected with crushing and sizing.

polyethylene technology were a few hundred thousand. Consequently, either of these technologies will produce a very large return on the DOE investment if deployed for stabilizing even a small portion of the DOE's depleted uranium.

Table 1. UF_6 Inventory Disposal Cost Comparison Between Grout, Polyethylene, and DUAGG Aggregate Stabilized Depleted Uranium Oxide[1]

	Grout	Poly	DUAGG
Estimated Loading Fraction of U_3O_8	25 %	90%	90%[2]
Container Bulk Loading Efficiency	95%	95%	60%
Total Mass Stabilized Material Shipped (Million MT)	2.77	0.988	0.454
Total No. of 55-gal Containers (millions)	5.48	1.05	.39
Total Shipments	144,344	43,086	33,307
Cost Estimate			
Processing Costs (millions)	$489	470	$285
Container Costs (millions)	$130	$24.8	$22.7
Transportation Cost (millions)	$650	$194	$150
Disposal Fee @ $8.50/ft^3 (millions)[3]	$324	$62	$28
Total Costs (millions)	$1,592	$751	$486

1. Starting material is assumed to be 550,000 MT of UF_6 that is converted to U_3O_8. Conversion costs from UF_6 to U_3O_8 are not included in above costs.
2. U_3O_8 converted to UO_2 in manufacturing process
3. Nevada Test Site disposal fee May 1998

COST ESTIMATE UNCERTAINTIES

Processing cost is the most significant uncertainty for all of the technologies evaluated. Both DUAGG and polyethylene processing are likely to be somewhat more costly than grout stabilization. Other costs such as disposal fee, transportation cost, and container cost are much more certain. From Table 1, it can be observed that the *processing cost* of polyethylene micro encapsulation and DUAGG aggregate could increase by more than 5 and 6 times, respectively, before their total predicted disposal costs would equal that of grout stabilization. Consequently, the cost reductions associated with the basic volume advantage offered by these new technologies should be retained despite processing cost uncertainties.

Table 2. Cost Model Assumptions

	Grout	Poly	DUAGG
Container Cost ($)	25	25	50[a]
Container Bulk Loading Efficiency	95%	95%	60%
Weight Per Container (kg)	505	787	1331
Transportation Cost Per Truck-Mile	1.80		
Loaded Truck Weight (kg/lb)	18,144 / 40,000		
Trip Distance (miles)	2500		
Reagent Cost $/lb-UOx	0.26	0.24	0.04
Processing Cost $/lb-UOx	0.25	0.25	0.25
Disposal Fee / ft^3	$8.50		

a. Container cost doubled to reflect need for stronger container to contain heavier load. Cost doubling impacts final cost by $0.03 per kg-U

IV. ENVIRONMENTAL BENEFITS

Present day practices at some DOE low-level disposal sites allow the direct disposal of depleted uranium without stabilization (At the Nevada Test Site, depleted uranium must be either stabilized to eliminate the fines or placed in an overpacked container or other secure packaging – Reference 12). From a heavy metal chemical toxicity hazard perspective, uranium metal is only slightly less hazardous than lead (Reference 13). Sandia has conducted performance assessments to determine the safety of radioactive material disposal at its low-level waste sites. Depleted uranium, because of its essentially infinite lifetime, was one of the most limiting radioactive isotopes for the groundwater pathway in wet disposal sites (Reference 14).

Thus, leaching characteristics of the uranium may influence the future environmental risk at a disposal site and disposal forms may require further evaluation. Disposal waste forms with lower leaching characteristics reduce both future environmental risk and economic risk associated with potential groundwater cleanup. Table 3 shows some comparative leaching results for various chemical forms of uranium.

These data were obtained from materials used or manufactured at the Starmet CMI facility in Barnwell, SC. The environmental benefits of DUAGG as a disposal

Table 3. Comparative Leach Test Results for Depleted Uranium Subjected to EPA TCLP Testing.	
Uranium Form	Concentration in Leachate (mg-U/liter)
DUAGG	4
U_3O_8	420
UF_4	7367
UO_3	6900
The UO_3 is from the DOE Savannah River Site and was recovered from reprocessing. The U_3O_8 and DUAGG were manufactured at Starmet CMI from SRS UO_3. The UF_4 was converted from UF_6.	

waste form over other forms of uranium is clearly evident in this data.

Since EPA regulations require lead stabilization to meet very stringent standards for leach resistance (0.37 mg/liter), it can be expected that future requirements for depleted uranium stabilization might parallel that of lead. These new technologies, DUAGG aggregate combined with a recycle product or as a waste form and polyethylene encapsulation both provide a superior leach resistance compared to direct disposal of uranium materials. Thus, if future stabilization standards are invoked for depleted uranium, the required technology is ready for deployment.

V. CONCLUSIONS

Advanced technologies developed through the DOE OST programs have offered new approaches to using or disposing of depleted uranium. These approaches, if deployed, provide both cost savings and improved environmental performance. The technologies provide new materials for designing nuclear shielding for spent fuel and high level waste. Alternate waste package design options utilizing a DUCRETE shielded overpack for the Yucca Mountain repository have been identified. In addition, more environmentally stable waste forms have been developed that offer a projected lower cost of disposal while at the same time, reduce the future environmental risk from leaching of uranium.

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